Distribution of Pesticides and Polycyclic Aromatic Hydrocarbons in House Dust as a Function of Particle Size

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House dust is a repository for environmental pollutants that may accumulate indoors from both internal and external sources over long periods of time. Dust and tracked-in soil accumulate most efficiently in carpets, and the pollutants associated with dust and soil may present an exposure risk to infants and toddlers, who spend significant portions of their time in contact with or in close proximity to the floor and who engage in frequent mouthing activities. The availability of carpet dust for exposure by transfer to the skin or by suspension into the air depends on particle size. In this study, a large sample of residential house dust was obtained from a commercial cleaning service whose clients were homeowners residing in the Raleigh-Durham-Chapel Hill (Research Triangle) area of North Carolina. The composite dust was separated into seven size fractions ranging from < 4 to 500 μm in diameter, and each fraction was analyzed for 28 pesticides and 10 polycyclic aromatic hydrocarbons (PAHs). Over 20% of the fractionated dust sample consisted of particles < 25 μm in diameter. Fourteen pesticides and all 10 of the target PAHs were detected in one or more of the seven size-fractionated samples. Sample concentrations reported range from 0.02 to 22 μg/g; the synthetic pyrethroids *cis*- and *trans*-permethrin were the most abundant pesticide residue. The concentrations of nearly all of the target analytes increased gradually with decreasing particle size for the larger particles, then increased dramatically for the two smallest particle sizes (4–25 μm and < 4 μm). *Key words*: dust composition, dust exposure, house dust, PAH, particle size, pesticides, polycyclic aromatic hydrocarbons. *Environ Health Perspect* 107:721–726 (1999). [Online 29 July 1999]
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House dust is a repository of pesticides and other chemicals used indoors or tracked in from outdoors. Pesticides become associated with house dust primarily through interior use of pest control formulations, intrusion of vapors from foundation and crawl-space treatments, and track-in of lawn and garden chemicals. Polycyclic aromatic hydrocarbons (PAHs) derive from indoor sources such as combustion, cooking, and smoking, as well as from track-in of contaminated yard soil or residues from garage floors. Once indoors where they are protected from environmental degradation, pollutants associated with dust persist for long periods, particularly if the dust is embedded in carpets. Starr et al. (*1*) were the first to report on pesticides in house dust. They analyzed dust from the homes of farmers and pesticide formulators in Colorado and found organochlorine pesticides in the 10–100 μg/g concentration range. Shortly thereafter, Davies et al. (*2*) reported that concentrations of pesticides in house dust were 15–50 times higher than those in the soils surrounding dwellings on the Caribbean Island of Bimini. They also found that air levels of 4,4′-DDT in these homes were elevated for 30 min after sweeping, and estimated that exposure to DDT in house dust exceeded that received from the diet. Lewis et al. (*3*) reported that the concentrations of pesticides associated with dust vacuumed from carpets in middle-income North Carolina homes with small children were 10–100 times higher than those found in the surface soils surrounding the house, and suggested that ingestion of house dust by small children through mouthing may represent an important contribution to their daily intake of pesticide residues. Likewise, Simcox et al. (*4*) obtained similar results from both agricultural and nonagricultural households in Washington State. PAH concentrations in house dust are also substantially higher than in yard and foundation soil (*5*). Recently, the National Cancer Institute (NCI), which is investigating a possible association between cancer risks and pesticides in house dust, found 19 of 26 pesticides, 10 of 10 PAHs, and 5 of 6 polychlorinated biphenyls targeted in house dust samples collected from 15 homes (*6*).

The translocation of pesticides and PAHs from the outdoors to the indoors and their redistribution indoors is a function of the sizes of the soil and house dust particles with which they are associated. The availability of dust-associated pollutants for respiratory exposure or ingestion by mouthing of hands, toys, or other objects likewise depends on particle size. Inhalable (< 10 μm) and respirable (< 2.5 μm) particles constitute the greatest risks for airborne particle-associated pesticide residues. Camann et al. (*7*) found strong and statistically significant correlations between the indoor air and carpet dust concentrations of several pesticides. Resuspension of 2,4-dichlorophenoxyacetic acid (2,4-D) residues tracked indoors from lawn applications was recently reported to be the major source of the herbicide in indoor air (*8*). Whereas larger particles are of concern for ingestion exposure, the efficiency of retention to the skin and the bioavailability of pollutants associated with dust and soil may be dependent on particle size (*9*). To better estimate the potential exposure to dust-associated pollutants, therefore, it is important to understand how such pollutants are distributed with respect to particle size. To this end, a large composite sample of residential house dust was obtained, fractionated by sieving and cyclone separation, then analyzed for a number of pesticides and PAHs commonly reported in house dust.

Methods

Four vacuum cleaner bags containing household dust were procured in November 1996 from a professional residential cleaning service located in the Raleigh-Durham-Chapel Hill (Research Triangle) area of North Carolina. The procurement was ancillary to a joint effort between the U.S. Environmental Protection Agency (EPA) and the National Institute of Standards and Technology (NIST) to obtain a representative sample for the development of a standard reference

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material for indoor dust. The collected dust was vacuumed from approximately 25 middle-class homes located in suburban neighborhoods in a nonindustrial nonagricultural area. The typical home was two-stories tall with a 185-m^2 average floor area, of which approximately 80% was carpeted. The homes typically did not have basements and were equipped with forced-air ventilation, gas heating units, and electric cook stoves. Commercial upright vacuum cleaners with unlined paper collection bags were used (empty-bag trapping efficiency of 65–75% for 2–4 µm particles at 1.7 m^3/min) and vacuuming was usually completed in 30–40 min per home.

The total gross weight of the sample (including the paper collection bags) was 6.35 kg. Each vacuum bag was opened and the contents were sieved separately to a particle size of < 2 mm with a Tyler Ro-Tap Sieve Shaker (Fisher Scientific, Pittsburgh, PA). The four sieved fractions were combined and sieved once again to achieve a coarse material consisting of particles < 2 mm in diameter and weighing a total of 2.72 kg. Because the dust was ultimately to be used in studies to determine the efficiency of transfer of the fractionated dust to human skin, it was sterilized by γ-irradiation at 2.5 mrad for 6 hr (Neutron Products, Inc., Dickerson, MD) before fractionation. Replicate analyses of aliquots taken from the coarse dust sample before and after γ-irradiation showed no significant concentration differences for any of the target analytes.

A 1.81-kg aliquot of the sterilized coarse dust sample (< 2 mm) was processed by Powder Technology, Inc. (Burnsville, MN), which used an air classifier to separate dust particles on the basis of aerodynamic diameter. The stepwise fractionation process involved screening the coarse sample at 35 mesh (> 500 µm) to remove larger particles that might plug the separation equipment. The 1.57-kg fraction resulting from that screening was then classified sequentially in two runs of the air classifier process to produce a nominal 98% < 25 µm fraction. Particle size analysis of this fraction revealed a tailing effect, with the 50% cumulative volume cut point at 28 µm (geometric diameter) and the 90% cut point at 106 µm. The size distribution data are shown in Figure 1A. All of the original sample material was accounted for by weighing each of the separate sample fractions.

The 1.16-kg dust sample remaining from the two air classifier runs after removal of the nominal < 25-µm fraction was sieved using the Ro-Tap apparatus to produce five additional size fractions, < 53 µm (-270 mesh), 53–106 µm, 106–150 µm, 150–250 µm, and 250–500 µm (+ 60 mesh). The small particle fraction from the second air classifier run constituted a sixth fraction made up of particles smaller than 25 µm.

To determine the concentrations of pollutants associated with respirable particles, a portion of the < 25-µm fraction was processed in 500-mg aliquots in an air classifier made up of a fluidized bed aerosol generator (FBAG), a 3.5-µm cyclone (Model URG-2000-30EC, URG, Carrboro, NC), and a 47-mm particle filter pack (Model URG-2000-30F). The FBAG was constructed by Peters and Vanderpool (*10*) based on a design by John and Wall (*11*). The FBAG uses 100-µm bronze beads mixed with each 500 mg dust charge to deagglomerate the sample with only minimal alteration of the original size distribution. To achieve a nominal 3.5-µm cut point with the cyclone, the FBAG was operated under positive pressure with prepurified dry nitrogen at a flow rate of 30 L/min. Dust particles resuspended by the FBAG were transported through a 90° bend and entered the cyclone inlet. In theory, particles with aerodynamic diameter larger than the 3.5-µm cut point dropped into a receiver cup, whereas particles smaller than the cut point were collected on a 47-mm quartz fiber filter at the exit of the apparatus. The overall efficiency of the dust aerosolization process and size separation was low. Approximately 11.5 g of the nominal < 25-µm fraction was processed to yield 3.74 g dust in the 4–25 µm size range and 0.124 g nominal < 4 µm dust, which was collected on a total of 12 filters. The remaining dust was lost to the walls of the apparatus or remained in the FBAG.

It is assumed that particle losses in the FBAG apparatus were independent of the particle surface loadings of pesticides and PAHs. In the unlikely event that surface loadings approach monolayer coverage, however, the sticking probabilities of the loaded particles may be altered, creating the potential for bias in the reported results; i.e., particles with near-monolayer loadings may have a higher or lower probability of being lost in the FBAG as compared to particles with low surface loadings. We have not confirmed such a bias nor has there been any attempt in this paper to correct the data for such a bias.

Verification of the size distribution in the < 4-µm-size fraction was performed by automated scanning electron microscopy (SEM). To this end, a polycarbonate membrane (1.0-µm pore) filter (Poretics Corp., Livermore, CA) lightly coated with mineral oil to enhance particle retention was used. The FBAG was again operated for 5 min, but the dust charge was reduced by a factor of 10 to produce light loading on the filter. SEM analysis of the coated filter determined the sizes of 3,820 particles. The resulting size distribution (Figure 1B) showed that the effective 50% cumulative volume cut point was 3.8 µm; 90% of the particles by volume had diameters < 6.6 µm.

All dust samples were stored under refrigeration (approximately 5°C) after fractionation.

Sample extraction. For the coarse dust sample and the six size-fractionated dust samples, approximately 2.0 g of each sample was spiked with an internal surrogate standard (*p*-terphenyl-d_{14}; Ultra Scientific, North Kingstown, RI) and then Soxhlet-extracted with 150 mL 6% diethyl ether in *n*-hexane for approximately 16 hr (*12*). The volume of each extract was reduced to 10 mL by a Kuderna-Danish concentrator. A 1-mL aliquot of each extract was passed through an 8-mm (i.d.) × 6-cm 100–200 mesh Florisil column (Supelco, Inc., Bellefonte, PA), and the final volume of each cleaned-up extract was adjusted to 2 mL. For the coarse and fine dust fractions generated from the FBAG/cyclone process, the same general extraction procedure was followed with the following exceptions. The 3.74-g coarse dust fraction was split into two nearly equal samples that were each extracted with 150 mL of solvent. The fine dust fraction (< 4 µm) collected on 12 quartz-fiber filters was extracted in two six-filter lots with 150 mL of solvent used for each lot. The extracts were reduced directly to a final volume of 2 mL without performing the Florisil column

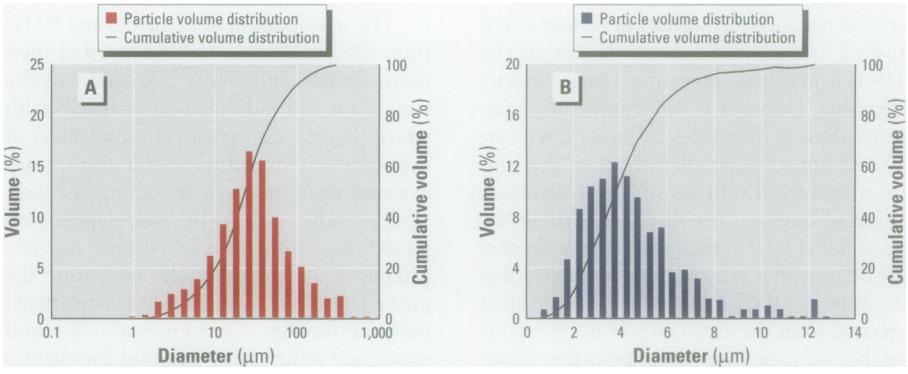

Figure 1. Particle volume distribution and cumulative volume distribution for (*A*) the nominal < 25-µm fraction and (*B*) the nominal < 4-µm fraction.

cleanup procedure. This method did not recover acid herbicides such as 2,4-D.

Gas chromatography/mass spectrometry (GC/MS) analysis. The dust sample fractions were analyzed for 28 neutral-extractable pesticides and 10 PAHs by capillary gas chromatography on a 0.25-mm (i.d.) × 30-m DB-5.625 column (J & W Scientific, Folsom, CA) coupled with a Fisons VG-MD800 mass spectrometer (Fisons, Danvers, MA) operated in the selected ion monitoring mode. The GC column temperature was programmed from 60°C (5 min hold) to 295°C at 15°C/min (with an intervention of 3 min at 200°C). Ionization was by 70 eV electron impact. Quantitative analyses were based on a five-point standard calibration curve and internal standards. Continuing calibration was performed using the mid-level standard. Extracts exhibiting target analyte concentrations above the calibration range were diluted and reanalyzed to bring the concentration within the range of the calibration curve. Detection limits were approximately 0.02–0.1 µg/g for pesticides and 0.02–0.05 µg/g for PAHs.

Quality assurance. The quality of the data derived from this study was assessed primarily by reviewing the data that resulted from performing all sample analyses in duplicate and by strict adherence to the procedures outlined in written protocols for performing the sample extraction and GC/MS analysis of the fractionated dust samples. Data quality was assessed for each sample analysis sequence by examining the percent relative standard deviation (RSD) of the relative response factors in the initial calibration curve for the surrogate and for each of the target analytes. In all cases the RSD was < 20% for the surrogate and target analytes. In addition, the percent differences were also compared for the continuing calibration standards with the initial calibration curve and were generally < 25%. Laboratory solvent blanks were routinely analyzed and no interferences were observed. Surrogate recoveries for each sample were reported and monitored for any indication of problems with the sample extraction procedure.

For the paired sample analyses, the average percent difference was calculated for each target analyte that was detected in both samples. A surrogate recovery standard, *p*-terphenyl-d_{14} (Ultra Scientific), was added to each dust sample fraction prior to extraction, and the percent recovery of that compound was determined for every sample analyzed. Previous studies demonstrated the efficiency of this extraction technique for all of the target analytes and the adequacy of *p*-terphenyl-d_{14} as the recovery surrogate (*11*). Solvent blanks were routinely analyzed along with each of the three sample sets that were processed, and in every case there were no target analytes detected in the blanks.

In all cases except those results from samples that were detected at or near the detection limit of the method, the reproducibility of the analytical process was acceptable. In all, 23 target analytes were detected in a minimum of 5 paired samples from the set of 10 paired samples that were analyzed. Of 13 pesticides and 10 PAHs detected, the mean and median values calculated for the average percent difference for sample pairs were always < 30% (mean differences ranged from 5.2 to 28.3%; median differences from 4.8 to 22.5%). Surrogate recovery data were good, with mean and median recoveries of 96.3 and 95%, respectively.

Results

Initial sieving of the vacuum bag contents to remove hair, fibers, and other particles > 2 mm in diameter resulted in the exclusion of 3.63 kg of material, or 57% of the 6.35 kg initial weight of the original sample, including that of the four vacuum bags. The remaining 2.72 kg of coarse dust was separated into eight discrete size-fractionated samples. Figure 2 breaks down the eight sample fractions by weight and weight percent based on the weight of the coarse dust sample. The largest fractions consisted of dust in the 53–106 µm and < 25 µm size ranges (22.7 and 20.7%, respectively, of the coarse dust). The estimated weight percent of dust in the respirable range was 0.7% based on the portion of the suspended < 25-µm fraction collected with the 3.5-µm cut-point inlet. An exact mass balance could not be determined because of deposition of suspended dust on the walls of the FBAG assembly. As much as 25–35% of the dust in the 2–4-µm size range may have been lost during collection by penetration of the vacuum bags (*13*). There may also have been additional losses of fine particles due to adherence to the walls of the vacuum bags.

The < 2-mm coarse fraction and seven subfractions of household dust were analyzed in duplicate by GC/MS for the target analyte list of 28 pesticides and 10 PAHs. Duplicate analysis of samples of the processed coarse dust before and after γ-irradiation assured that there were no detectable losses of analytes due to sterilization. The mean percent recovery of the internal standard (*p*-terphenyl-d_{14}) for 20 samples analyzed was > 96%. The mean percent difference for the 201 replicate analytical results calculated for the individual target analytes fell between 5 and 28%.

A total of 13 of the pesticides and all 10 of the PAHs on the target list were detected in one or more of the sample pairs analyzed. The pesticides ranged in concentration from 0.02 to 21.9 µg/g; the highest levels detected were those for *cis*- and *trans*-permethrin. The PAHs ranged in concentration from 0.04 to 2.03 µg/g, with chrysene and benzo[*b*]fluoranthene present at the highest levels. The data presented in Table 1 represent the mean values of the duplicate analyses for each of the samples. None of the other 15 targeted pesticides (alachlor, aldrin, atrazine, captan, chlorothalonil, dacthal, diazinon, dicofol, dieldrin, folpet, heptachlor, hexachlorobenzene, lindane, malathion, and resmethrin) were detected in the coarse dust sample or in any of the size fractions. The detection limits for the undetected pesticides ranged from 0.1 to 0.2 µg/g for the < 4-µm fraction and from 0.02 to 0.07 µg/g for the other fractions. Results for the coarse dust sample (< 2 mm) most closely matched those for the 53–106 µm fraction, which was the largest weight proportion (22.7%).

The commonly applied pyrethroid insecticides *cis*- and *trans*-permethrin were present in the highest concentrations. These pesticides are also frequently incorporated into new carpeting at the time of manufacture. Two other common household insecticides, chlorpyrifos and carbaryl, were also present in relatively high concentrations. The concentrations of all analytes detected generally increased with decreasing particle size, suggesting that they were primarily attached to the surfaces of the particles (as opposed to being absorbed or trapped within the dust particles). The rate of concentration increase for most analytes escalated on particles below 100 µm, especially in the case of PAHs. In all cases except that for coronene, concentrations were highest on the smallest particles (< 4 µm). The most pronounced concentration increases for pesticides occurred between the 25–53 µm and 4–25 µm fractions and the 4–25-µm and < 4-µm fractions. For PAHs, the largest increase in concentration always occurred between the 4–25-µm and < 4-µm fractions. DDT and its degradation product DDE, *o*-phenylphenol, and dibenzo(*a*,*e*)pyrene were only detected on particles < 100 µm. The concentration distributions of eight pesticides and eight PAHs are presented graphically in Figures 3 and 4.

The concentrations of several of the pesticides and PAHs detected show some

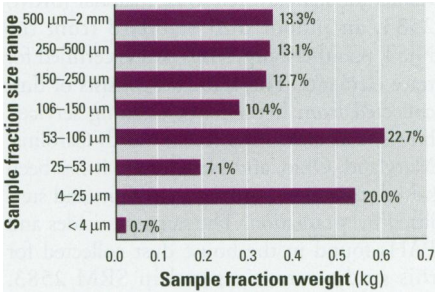

Figure 2. Weight of separated dust fractions and percent of total coarse (< 2 mm) dust weight for each size fraction.

Table 1. Analysis of size-fractionated household dust for neutral-extractable pesticides and polycyclic aromatic hydrocarbons.

	Analyte concentrations (µg/g) by size fraction								
Target analytes	Coarse dust (< 2 mm)	250–500 µm	150–250 µm	106–150 µm	53–106 µm	25–53 µm	4–25 µm	< 4 µm	SRM 2583[a] (< 100 µm)
Bendiocarb	0.36	0.11	0.13	0.14	0.18	0.16	0.71	1.43	< 0.08
Carbaryl	0.87	0.19	0.49	0.47	0.65	0.39	2.56	4.61	4.23
α-Chlordane	0.09	< 0.05	0.04	0.06	0.09	0.12	0.24	0.49	0.07
γ-Chlordane	0.16	< 0.05	0.07	0.10	0.15	0.19	0.34	0.65	0.12
Chlorpyrifos	0.54	0.16	0.22	0.29	0.44	0.56	1.05	4.52	0.54
4,4′-DDE	0.02	< 0.06	< 0.06	< 0.06	0.02	0.02	< 0.04	< 0.13	< 0.11
4,4′-DDT	0.04	< 0.05	< 0.05	< 0.05	0.04	0.05	0.13	0.71	< 0.10
Diazinon	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.04	< 0.13	0.90
Dichlorvos	< 0.07	< 0.07	< 0.07	< 0.07	< 0.07	< 0.07	< 0.05	0.15	< 0.14
Heptachlor	< 0.16	< 0.16	< 0.16	< 0.16	< 0.16	< 0.16	< 0.13	< 0.40	0.30
Methoxychlor	0.56	0.12	0.21	0.31	0.57	0.74	0.68	1.00	0.10
cis-Permethrin	5.32	1.34	2.26	2.96	4.67	5.83	12.08	15.17	1.74
trans-Permethrin	7.29	1.50	2.73	3.70	6.21	7.15	17.45	21.87	2.50
o-Phenylphenol	0.07	< 0.06	< 0.06	< 0.06	0.05	0.04	0.18	1.20	0.34
Propoxur	0.08	0.04	0.05	0.04	0.05	0.03	0.18	0.38	0.16
Benz[*a*]anthracene[b]	0.23	0.10	0.09	0.12	0.21	0.22	0.30	0.97	0.88
Benzo[*b*]fluoranthene[b]	0.55	0.16	0.17	0.26	0.53	0.61	0.56	2.03	1.39
Benzo[*k*]fluoranthene[b]	0.30	< 0.05	0.05	0.08	0.16	0.19	0.18	0.92	1.46
Benzo[*ghi*]perylene	0.34	0.08	0.09	0.16	0.38	0.48	0.38	1.33	1.09
Benzo[*a*]pyrene[b]	0.28	< 0.05	0.05	0.07	0.16	0.18	0.27	1.08	0.97
Chrysene[b]	0.41	0.14	0.15	0.21	0.41	0.47	0.65	1.67	1.72
Coronene	0.37	0.07	0.08	0.14	0.36	0.46	0.11	0.35	1.22
Dibenz[*a,h*]anthracene[b]	0.08	< 0.05	< 0.05	0.04	0.08	0.10	<0.04	0.22	0.22
Dibenzo[*a,e*]pyrene	0.10	< 0.05	< 0.05	< 0.05	0.08	0.11	0.09	0.30	0.25
Indeno[1,2,3-*cd*]pyrene[b]	0.34	0.08	0.08	0.13	0.30	0.35	0.30	1.25	0.99

[a]National Institute of Standards and Technology (Gaithersburg, MD) indoor dust standard reference material. [b]Classified B2 probable human carcinogens by the U.S. Environmental Protection Agency (*16*).

evidence of bimodal distribution. The distribution patterns of carbaryl, bendiocarb, and methoxychlor, unlike most of the other principal pesticides detected, exhibited a dip in concentration followed by a continued increase with decreasing particle size. Carbaryl may still be used by homeowners for indoor flea control and on pets, although current label instructions restrict its use to outdoor applications. It is most frequently applied as a powdered formulation or dust on talc, which may account for its distribution differing from that of most of the other pesticides. Bendiocarb is used to control ants, cockroaches, crickets, silverfish, wasps, etc. in houses, and is often applied as a dust or wettable powder. Methoxychor is a DDT analog that has been used on fruit trees, frequently in mixtures containing carbaryl and other insecticides. It is likely a track-in residue from past outdoor use.

Also shown in Table 1 are our analytical results for standard reference material (SRM) 2583, an indoor dust standard from the NIST (Gaithersburg, MD) that is certified for trace elements. This standard consists of dust collected from households, cleaning services, hotels, and motels located in North Carolina, Maryland, Ohio, and New Jersey. It has been sieved to remove particles > 100 µm and sterilized by γ-radiation. The same pesticides and PAHs found in the house dust collected for this study were measured in SRM 2583. Concentrations of PAHs in SRM 2583 were generally closer to those found in < 25 and < 4 µm fractions of our dust sample. This suggests either composition differences due to higher fossil fuel use in the northern states or due to a higher proportion of fine particles in the NIST standard. Two of the targeted pesticides found in the SRM (diazinon and heptachlor) were not detected in dust collected for this study. On the other hand, three of the pesticides found in the North Carolina dust sample (bendiocarb, DDT, and dichlorvos) were not detected in SRM 2583.

Discussion

The inverse proportionality of residue concentrations with dust particle size observed in this study clearly demonstrates that neutral-extractable pesticides and PAHs are found primarily on the surfaces of particles. Pesticide concentrations increased by 1.25- to 6.7-fold (mean 2.4-fold, median 2-fold) between the < 4 and 4–25 µm fractions and 1.2- to 3.9-fold (mean 2.8-fold, median 2.8-fold) between the 4–25 and 53–106 µm fractions. PAH concentrations increased more dramatically between the < 4 and 4–25 µm fractions (2.6- to 5.1-fold, mean 3.6-fold, median 4-fold), but their concentrations were about the same on the 4–25 and 53–106 µm fractions. If house dust was composed of perfect spheres with the same density and diameter, the total surface area of a given mass of 4 µm particles would be 6.25 times that of the same mass of 25 µm particles and 25 times that of 100 µm particles. House dust, however, is typically composed of soil, settled particulate matter, human and animal dander, insect parts, and other debris of varying shape, density, and porosity; thus, the concentrations of surface pollutants would not follow the same scale, even if they were uniformly distributed on the surfaces. Furthermore, pollutant concentrations would not be expected to be evenly distributed on the particles, especially if the particles and/or pollutants originated from different sources (e.g., interior use, air intrusion, or track-in). Also, each of the size fractions given in Table 1 and shown in Figures 3 and 4 includes many particle sizes of unknown relative mass distribution.

The major pesticide residues found in this study are similar to those reported by others. Camann and Buckley (*14*) reported detected values of *cis*- and *trans*-permethrins at median (maximum) concentrations of 1.75 (588) µg/g and 0.80 (299) µg/g, respectively; chlorpyrifos at 0.56 (324) µg/g; carbaryl at 0.40 (1,160) µg/g; bendiocarb at 0.36 (318) µg/g; methoxychlor at 0.59 (28) µg/g; and α- and γ-chlordane at 0.4 (15) µg/g and 0.46 (17) µg/g, respectively, in dust collected from 362 homes in nine northern and midwestern U.S. states and sieved to < 150 µm. However, these samples showed higher residues of older pesticides such as DDT (median 0.10, maximum 103 µg/g) and dieldrin (median 0.25, maximum 139 µg/g). Chlorpyrifos was reported by Simcox et al. (*4*) in household dust collected from 47 of 48 farm homes in the state of Washington in 1992 at a mean concentration of 0.43 µg/g (maximum 3,585 µg/g), as compared to 0.17 µg/g (maximum 483 µg/g) in 9 of 11 reference homes. Lewis et al. (*3*) found chlorpyrifos at 1.6 µg/g (maximum 3.1 µg/g), chlordane at 1.8 µg/g (maximum 2.8 µg/g), heptachlor at 0.27 µg/g (maximum 0.72 µg/g), dieldrin at 0.29 µg/g (maximum 1.0 µg/g), and pentachlorophenol at 0.83 µg/g (maximum 3.3 µg/g) in carpet dust from nine homes in the same geographic area as this study. Pentachlorophenol, the most frequently detected pesticide in the Lewis et al. (*3*) study, was not targeted in the current investigation nor in that of Camann and Buckley (*14*). In all three of the aforementioned studies (*3,4,14*), carpet dust was collected with the high-volume surface sampler (HVS3, CS_3, Inc., Bend, OR) vacuum sampler (*3*), which has a nominal cut point of 5 µm for collected dust. A side-by-side comparison of the HVS3 and a conventional upright vacuum cleaner showed that both collected particles down to at least 0.2 µm and that the HVS3 was more efficient at collecting particles < 20 µm (*34*). Because pesticide and PAH concentrations increase rapidly on particles < 25–50 µm, analytical results for dust collected with household vacuum cleaners might be expected to be lower than those obtained with the HVS3. No significant differences in the concentrations of pesticides and PAHs were found by

the NCI in house dust collected with the HVS3 and the concentrations in dust taken from the bags of vacuum cleaners used in the homes (*6*); however, a recent EPA study of nine day-care centers yielded higher results for pesticides and PAHs in dust collected in standard vacuum cleaner bags in most cases (*15*). In the NCI study (*6*), the HVS3 sample was collected from carpets throughout the house. In contrast, EPA investigators collected the HVS3 sample from a single room on one day, whereas the bag sample was taken from the home or facility vacuum cleaner and represented dust collected over an unknown period of time and from multiple locations within the building. Consequently, concentration differences in the two types of samples reported by the NCI and the EPA may have reflected a lack of spatial and temporal homogeneity of the dusts.

The total concentration in the coarse house dust sample of the seven B2 PAHs, which are classified by the EPA as probable human carcinogens based on animal studies (*16*), was 2.21 µg/g. This value is similar to the mean concentration of 1.73 µg/g for the same analytes measured in dust collected

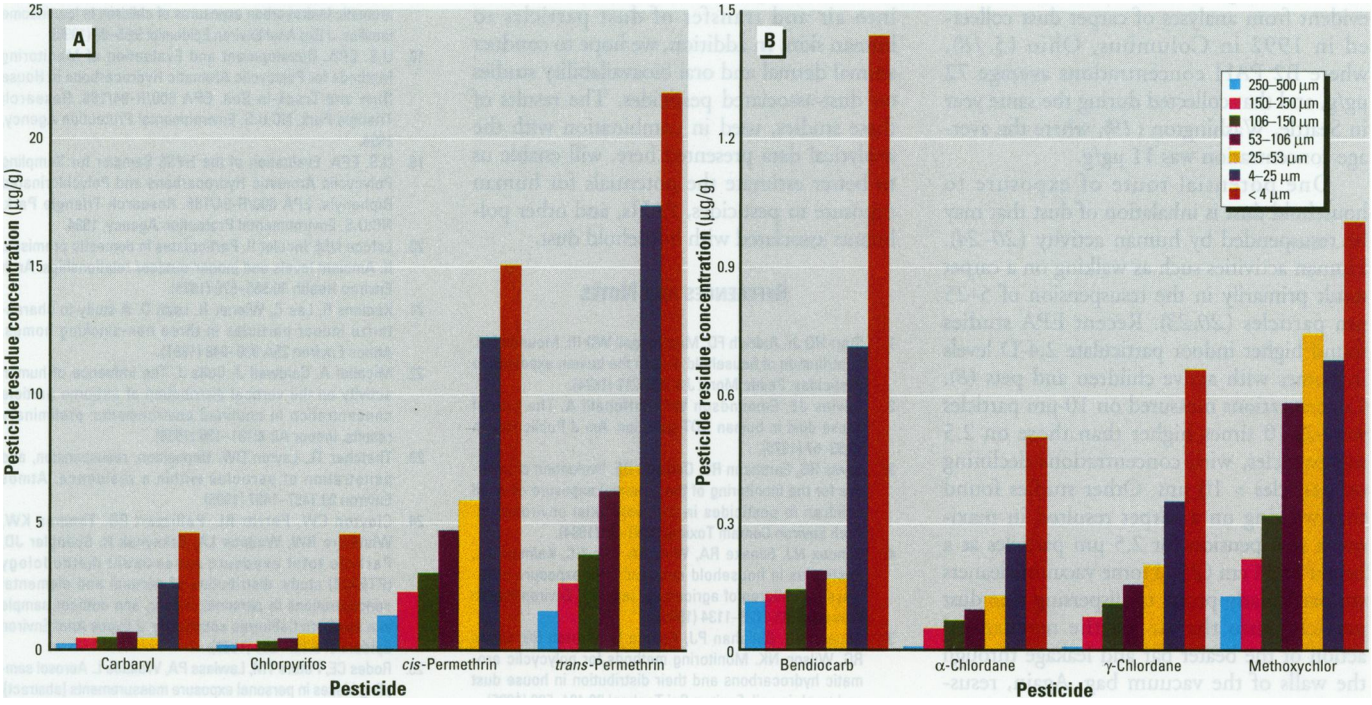

Figure 3. Concentrations in micrograms per gram of selected pesticides in house dust by size fraction.

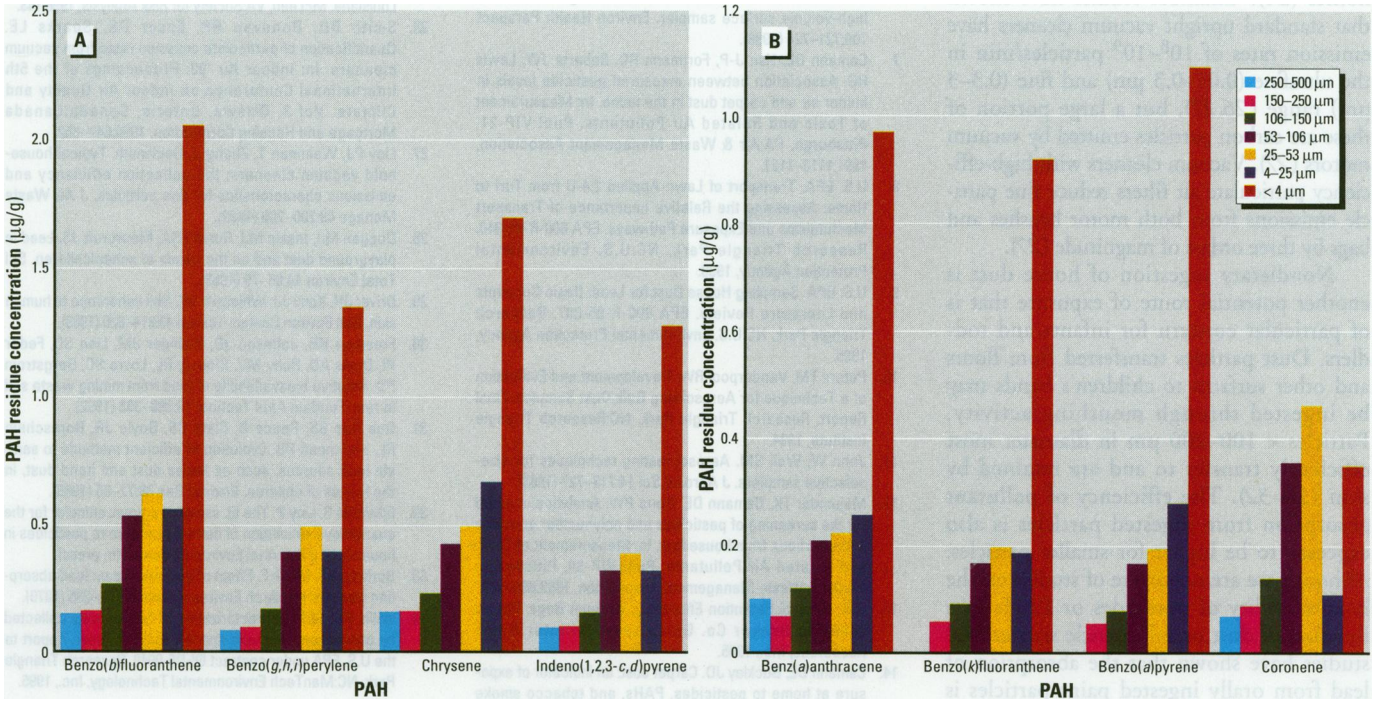

Figure 4. Concentrations in micrograms per gram of selected polycyclic aromatic hydrocarbons (PAHs) in house dust by size fraction.

from low-income homes in Durham, North Carolina, in 1994 (*17*). The sum of the B2 PAHs in SRM 2583 was substantially higher at 7.63 µg/g, perhaps reflecting geographic differences in PAH house dust levels between cities in southern and northern regions of the United States or the relative proportions of sources exposed to indoor tobacco smoking. The influence of fossil fuel use for heating on PAH house dust levels is evident from analyses of carpet dust collected in 1992 in Columbus, Ohio (*5,18*), where B2 PAH concentrations average 72 µg/g, and that collected during the same year in Seattle, Washington (*19*), where the average concentration was 11 µg/g.

One potential route of exposure to household dust is inhalation of dust that may be resuspended by human activity (*20–24*). Human activities such as walking on a carpet result primarily in the resuspension of 5–25 µm particles (*20,23*). Recent EPA studies found higher indoor particulate 2,4-D levels in homes with active children and pets (*8*). Concentrations measured on 10-µm particles were 2–10 times higher than those on 2.5 µm particles, with concentrations declining on particles > 10 µm. Other studies found that walking on a carpet resulted in maximum resuspension for 2.5 µm particles at a height of 34 cm (*25*). Home vacuum cleaners are particularly prone to dispersing fine dust particles into the air via the mechanical action of the beater bar and leakage through the walls of the vacuum bag. Again, resuspended particles > 10 µm appear at the highest concentrations during vacuuming in homes (*24*). Chamber studies have shown that standard upright vacuum cleaners have emission rates of 10^8–10^9 particles/min in the ultrafine (0.01–0.3 µm) and fine (0.3–3 µm) range (*26,27*), but a large portion of these are carbon particles emitted by vacuum motors (*27*). Vacuum cleaners with high-efficiency particulate air filters reduce fine particle emissions from both motor brushes and bags by three orders of magnitude (*27*).

Nondietary ingestion of house dust is another potential route of exposure that is of particular concern for infants and toddlers. Dust particles transferred from floors and other surfaces to children's hands may be ingested through mouthing activity. Particles < 100–200 µm in diameter most efficiently transfer to and are retained by skin (*28–32*). The efficiency of pollutant absorption from ingested particles is also expected to be higher for smaller particles. Although we are not aware of studies on the bioavailability of pesticides or PAHs as a function of dust or soil particle size, animal studies have shown that the absorption of lead from orally ingested paint particles is significantly greater for smaller particles (e.g., < 50 µm) (*33*).

It is apparent that knowledge of the distribution of pollutants on house dust and soil is important to the understanding of human exposure. Studies are currently underway in our laboratory to determine the dependence of soil track-in efficiency on particle size, as well as to better characterize resuspension of dust from carpeted and vinyl-covered floors into air and transfer of dust particles to human skin. In addition, we hope to conduct animal dermal and oral bioavailability studies on dust-associated pesticides. The results of these studies, used in combination with the analytical data presented here, will enable us to better estimate the potentials for human exposure to pesticides, PAHs, and other pollutants associated with household dust.

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